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# Fossil and non-fossil sources of organic carbon (OC) and elemental carbon (EC) in Göteborg, Sweden

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## Abstract

Particulate matter was collected at an urban site in Göteborg (Sweden) in February/March 2005 and in June/July 2006. Additional samples were collected at a rural site for the winter period. Elemental carbon (EC), organic carbon (OC), water-insoluble OC (WINSOC), and water-soluble OC (WSOC) were analyzed for  $^{14}\text{C}$  in order to distinguish fossil from non-fossil emissions. As wood burning is the single major source of non-fossil EC, its contribution can be quantified directly. For non-fossil OC, the wood burning fraction was determined independently by levoglucosan and  $^{14}\text{C}$  analysis and combined using Latin-hypercube sampling (LHS). For the winter period, the relative contribution of EC from wood burning to the total EC was >3 times higher at the rural site compared to the urban site, whereas the absolute concentrations of EC from wood burning were comparable at both sites. Thus, the urban site is substantially more influenced by fossil EC emissions. For summer, biogenic emissions dominated OC concentrations most likely due to secondary organic aerosol (SOA) formation. During both seasons, a more pronounced fossil signal was observed for Göteborg than has previously been reported for Zurich, Switzerland. Analysis of air mass origin using back trajectories suggests that the fossil impact was larger when local sources dominated, whereas long-range transport caused an enhanced non-fossil signal. In comparison to other European locations, concentrations of levoglucosan and other monosaccharide anhydrides were low for the urban and the rural site in the area of Göteborg during winter. The comparison of summer and winter results provides insight into the annual cycle of anthropogenic vs. biogenic contributions to the atmospheric aerosol.

## 1 Introduction

Airborne particulate matter (PM) influences the radiation budget directly by scattering of sunlight and indirectly by cloud formation. Furthermore, it causes respiratory as well as cardiovascular diseases (Knaapen et al., 2004). The carbonaceous fraction, which

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is a main constituent of PM, contributes to these effects due to their particulate character (Jacobson et al., 2000; Kanakidou et al., 2005). Elemental carbon (EC), which is optically absorptive and highly polyaromatic, has enhanced negative repercussions on human health (Knaapen et al., 2004). On the other hand, reactive components of organic carbon (OC), the lighter fraction of the carbonaceous aerosol, take part in atmospheric-chemical transformations (Tsigaridis et al., 2006). A large fraction of OC can be attributed to secondary organic aerosols (SOA), which is formed from gaseous precursors by atmospheric oxidation or oligomerization, whereas EC enters the atmosphere exclusively as a primary (i.e. direct particulate) emission (Jacobson et al., 2000; Kanakidou et al., 2005; Tsigaridis et al., 2006; Bond et al., 2007).

In spite of the importance of the carbonaceous aerosol, detailed apportionment and quantification of its sources is still difficult due to the large number of sources and the vast number of organic compounds associated with the aerosol (Kanakidou et al., 2005). In general, EC nearly completely originates from pyrolysis during incomplete combustion, e.g. due to traffic, industrial incineration, or biomass burning (Bond et al., 2007). The OC fraction includes primary particles and SOA originating from anthropogenic sources, e.g. from combustion processes in line with EC emissions. Besides those sources, primary biogenic emissions and SOA formation play an important role for the OC fraction, especially during summer. However, the actual production pathways and emission strengths of both, primary biogenic particles and SOA, are still poorly understood (Kanakidou et al., 2005). Additionally, numerous minor OC sources exist, which largely differ seasonally and locally. The above-mentioned sources all have complex emission and transformation regimes, which make inventory-based emission estimation challenging. Therefore, receptor models aim to re-construct the origin and emissions based on ambient concentrations of source-specific markers (e.g., organic compounds, inorganic tracers, or mass fragments of direct PM analyzing mass spectrometers) using model-derived or a priori established emission factors (Schauer et al., 1996; Hopke et al., 2006; Alfarra et al., 2007). This approach assumes that these emission factors are independent of changing emission conditions or chemical transfor-

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mations during atmospheric alteration. Whereas such assumptions are typically valid only under certain circumstances (e.g., close to emissions or for similar sites), they are much less problematic for many isotopic tracers. For carbonaceous particles, radiocarbon ( $^{14}\text{C}$ ) is such an isotopic tracer, by which fossil and non-fossil emissions can be distinguished (Currie, 2000; Lewis et al., 2004). With this technique, the contribution of biogenic and fossil OC for summer conditions (Szidat et al., 2004a, b) as well as the fraction of EC from residential wood burning during winter was directly quantified for several Swiss sites (Szidat et al., 2006; 2007). Furthermore, the combination of  $^{14}\text{C}$  analysis with other apportionment tools such as aerosol mass spectrometry (Alfarra et al., 2007), multi-wavelength aerosol light absorption measurements (Sandradewi et al., 2008a), organic tracer determination (Gelencsér et al., 2007), and aerosol modeling (Simpson et al., 2007) has recently gained new insights into sources of carbonaceous aerosols.

Levoglucosan is the most recognized molecular marker for tracing emissions of particulate matter from biomass burning. It is emitted in high concentrations, not present in the vapour phase, and predominantly associated with fine aerosols (Locker, 1988; Simoneit et al., 1999; Yttri et al., 2005). According to present knowledge it is not selectively removed from the atmosphere (Fraser and Lakshamanan, 2000). The stability of the levoglucosan/OC emission ratio for different burning conditions is somewhat controversial. Jordan and Seen (2005) stated that this ratio is stable for a wide range of burn rates, whereas many compilations of literature results (e.g., Szidat et al., 2006; Puxbaum et al., 2007; Sandradewi et al., 2008b) show quite variable relationships. However, levoglucosan enables detection of plumes from biomass burning and make it possible to study temporal variations of wood burning emissions on a sub-daily scale.

The present work employs  $^{14}\text{C}$  and levoglucosan determinations for two locations (urban and rural) in South-West Sweden in order to investigate the different source patterns of carbonaceous aerosols for this part of Scandinavia. Northern Europe provides a number of interesting conditions for investigating sources of carbonaceous aerosols.

1. Large forested areas are likely to give a greater proportion of biogenic to anthro-

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pogenic VOCs and aerosols than for other parts of Europe.

2. Wood-burning is quite common: one third of Swedish homes are completely or partially heated by burning of firewood (Hedberg et al., 2002). Glasius et al. (2006) stated that the number of wood-combustion appliances in Denmark has doubled during the last decade and demonstrated that wood combustion can result in local particle levels comparable to heavily trafficked streets.
3. Modeled SOA/OC ratios close to one are reported for Scandinavia both for summer and winter, which is on the regional scale substantially higher than for Central Europe (Simpson et al., 2007): In summer, the combination of high forest density and increased day length promote photochemical reactions of biogenic VOCs; in winter, low temperatures favor condensation of semi-volatile organic gases (Andersson-Sköld and Simpson, 2001; Simpson et al., 2007).

In this work, we present an assessment through  $^{14}\text{C}$  analysis of the contribution of fossil versus non-fossil carbon to ambient aerosols in and close to Gothenburg in South-West Sweden. Combined with the measurements of levoglucosan, this complementary  $^{14}\text{C}$  analysis provides a good estimate of the relative roles of biogenic VOC, anthropogenic wood-burning, and combustion sources to EC and OC. Furthermore, the comparison of summer and winter results yields insight into the seasonal cycle of anthropogenic vs. biogenic contributions to the atmospheric aerosol.

## 2 Methods

### 2.1 Particulate matter sampling

In February/March 2005 and June/July 2006, aerosol filter samples were collected for subsequent analysis of its carbonaceous content during the Göte-2005 campaign

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(Hallquist et al., 2008<sup>1</sup>) in the area of Göteborg, Sweden. Sampling during winter was conducted simultaneously at the urban site Femman and the rural EMEP site Råö (SE14), which is located 35 km south of Göteborg. During summer, sampling was conducted at the urban site only. The urban site is situated at an approximate height of 30 m above ground level in down-town Göteborg on top of the 7-floor-high department store Femman. This site is the main air quality monitoring site in Göteborg where routine measurements are conducted by the Environmental Office in Göteborg.

The aerosol was collected on pre-heated quartz fiber filters (Pallflex 2500QAT-UP) using different samplers, inlets, and air flows. For details see Table 1. During the winter campaign sampling was conducted using different inlets, i.e. PM<sub>10</sub> at Femman and PM<sub>2.5</sub> at Råö. These data are only fully comparable for conditions when the fine (particle diameters <2.5 μm) fraction dominates the organic load, while any occurring coarse organic fraction at Råö might influence this comparability. After sampling, filters were folded, wrapped in aluminum foil, packed into air-tight plastic bags, and stored at –20°C. During transportation, the filter samples experienced ambient temperatures for 48 h. Blank filters were treated in same way as the exposed filters without collection of aerosols.

## 2.2 Separation of carbonaceous particle fractions

The chemical separation of OC, WINSOC, and EC from the quartz fiber filters for <sup>14</sup>C measurement using a step-wise oxidation (Szidat et al., 2004c) was described elsewhere (Szidat et al., 2004a, 2006, 2007). In brief, OC is oxidized at 340°C in a stream of pure oxygen. For WINSOC analysis, water-soluble compounds are removed by water extraction of the filter. The remaining carbon on the filter is then treated as for OC

<sup>1</sup>Hallquist, M., Andersson, P. U., Boman, J., Emren, A., Hagström, M., Hasegawa, S., Janhäll, S., Langer, S., Ljungström, E., Nemitz, E., Olofsson, K. F. G., Pettersson, J. B. C., Synal, H.-A., Szidat, S., Shannigrahi, A. S., Svane, M., Thomas R., and Yttri, K. E.: Chemical characterisation of the urban aerosol during Göte-2005, in preparation, 2008.

separation. The level of WSOC is determined by subtraction of WINSOC from OC. EC is oxidized at 650°C after complete removal of OC and interfering water-soluble inorganic compounds, which is done by extraction with diluted hydrochloric acid and water followed by pre-heating at 390°C for 4 h. CO<sub>2</sub> evolving from OC, WINSOC, and EC is cryo-trapped and sealed in ampoules for <sup>14</sup>C measurement.

The concentrations of the carbonaceous fractions are determined using a modified procedure of that reported by Lavanchy et al. (1999). TC is quantified manometrically as CO<sub>2</sub> from the oxidation of the untreated filter at 650°C. For EC measurement, OC is eliminated by water extraction and pre-heating at 375°C during 40 min.

### 2.3 <sup>14</sup>C measurements

<sup>14</sup>C from the evolved CO<sub>2</sub> was analyzed in carbon amounts of 10–70 μg with accelerator mass spectrometry (AMS) at the PSI/ETH AMS facility, using two different techniques. For the majority of the samples collected during the winter campaign, CO<sub>2</sub> was reduced with a mixture of Co and Mn, forming filamentous carbon (Szidat et al., 2004c). This material served as the target material in the 500 kV pelletron AMS system, which is equipped with a MC SNICS cesium sputter ion source for solid targets (Synal et al., 2000). Due to isotopic fractionation effects and chemical contamination during the reduction step, correction of raw data is laborious and, sometimes, induces large uncertainties (Jenk et al., 2007). For three of the samples collected at Råö in winter (14–18 February, WINSOC and EC, and 18–25 February, WINSOC) and for all those collected in summer, CO<sub>2</sub> was mixed with He and transferred into a self-constructed cesium sputter gas ion source of the 180 kV mini-radiocarbon dating system MICADAS (Ruff et al., 2007). The latter method enables <sup>14</sup>C determinations with a higher reliability and less uncertainty compared to the former one and is only subjected to negligible chemical contamination. An intercomparison revealed that both methods produce consistent results. <sup>14</sup>C measurements are given in fractions of modern carbon ( $f_M$ ), which denote the <sup>14</sup>C/<sup>12</sup>C ratios of the samples related to that of the reference year 1950.

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According to Stuiver and Polach (1977), these values are decay-corrected for the time period between 1950 and the date of measurement.

## 2.4 Levoglucosan determination

Quantification of monosaccharide anhydrides (MA) (levoglucosan, mannosan and galactosan) in the aerosol filter samples was performed according to the method described in detail by Dye and Yttri (2005). In brief, punches ( $1.5\text{--}3\text{ cm}^2$ ) of the quartz fiber filter were soaked in tetrahydrofuran (2 ml) and subjected to ultrasonic agitation (30 min). The filter extract were filtered through a syringe filter ( $0.45\ \mu\text{m}$ ) to remove PM and filter parts. Each filter was extracted twice. The extracted volumes were pooled and evaporated to a total volume of 1 ml in a nitrogen atmosphere. Before analysis the sample solvent elution strength was adapted to the mobile phase by adding Milli-Q water (0.8 ml). The concentrations of MAs were determined using High Performance Liquid Chromatography (HPLC; Agilent 1100) in combination with HRMS-TOF (High Resolution Mass spectrometry Time-of-Flight; Micromass LCT) operated in the negative electrospray mode. Levoglucosan, mannosan, and galactosan were identified on the basis of retention time and mass spectra of authentic standards. The limit of quantification of the current method is 30 pg at a signal-to-noise ratio of 10.

## 2.5 Source apportionment of EC and OC

Source apportionment of fossil and wood-burning EC ( $EC_{\text{fossil}}$ ,  $EC_{\text{wood}}$ ), as well as fossil and non-fossil OC ( $OC_{\text{fossil}}$ ,  $OC_{\text{nonfossil}}$ ) is based on determination of EC, OC and TC concentrations and  $^{14}\text{C}$  isotopic ratios ( $f_M(\text{OC})$ ,  $f_M(\text{EC})$ ) (Szidat et al., 2006). EC is distinguished into  $EC_{\text{fossil}}$  and  $EC_{\text{wood}}$ . OC is separated into  $OC_{\text{fossil}}$  and  $OC_{\text{nonfossil}}$ , of which the second fraction comprises  $OC_{\text{bio}}$  and  $OC_{\text{wood}}$ . In order to differentiate between the latter two sources,  $OC_{\text{wood}}$  is estimated using two independent techniques, namely  $EC_{\text{wood}}$  and levoglucosan measurements using average emission ratios for residential wood burning in fireplaces (i.e.,  $(\text{OC}/\text{EC})_{\text{wood}}$  as given by Szidat et al. (2006)

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and  $(OC/lev)_{wood}$  as discussed in Sect. 3.4).  $OC_{bio}$  is determined by subtraction of  $OC_{wood}$  from  $OC_{nonfossil}$ . In order to allow for the multitude of possible combinations of parameters, an effective statistical approach known as Latin-hypercube sampling (LHS) was used, which is comparable to Monte Carlo calculations. Details of this procedure are given by Gelencsér et al. (2007). Ambient carbonaceous particles are assumed as a mixture originating from sources with pure isotopic signals. The following  $f_M$  figures have been applied for pure fossil, biogenic, and wood-burning sources:  $f_{M,fossil}=0$ ,  $f_{M,bio}=1.055\pm 0.015$ ,  $f_{M,wood}=1.16\pm 0.05$ . The values of the contemporary carbon sources ( $f_{M,bio}$  and  $f_{M,wood}$ ), which are elevated compared to the theoretical modern level of 1 due to the nuclear-bomb excess, are updated compared to earlier studies (Szidat et al., 2004a; 2006; 2007):  $f_{M,bio}$  for 2005/2006 was taken from the long-term time series of  $^{14}CO_2$  measurements at the rural site Schauinsland (Levin and Kromer, 2004; Levin et al., 2008),  $f_{M,wood}$  represents the integral signal of 30–50-year-old trees which were harvested in 2004 based on a tree growth model (Lewis et al., 2004; Mohn et al., 2008). For the source apportionment of the sub-fractions of OC,  $f_M(WINSOC)$  was measured directly and  $f_M(WSOC)$  was determined by concentration-weighted balancing of isotopic values of WINSOC and OC. These fractions were distinguished between fossil and non-fossil origin without further analysis of the latter portion due to highly uncertain WINSOC/OC emission ratios.

## 3 Results and discussion

### 3.1 Air mass origin

10-days FLEXTRA back trajectories (Stohl and Seibert, 1998) with a time resolution of 6 h were analyzed for the rural site Råö during the campaign. Due to the proximity of both locations and the simple topography of the landscape, aerosol contributions from long-range transport are assumed equal for the two sites with additional contributions of local sources, urban and rural, respectively. The borderline of regional sources is

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more complex due to local wind patterns in Göteborg area which may occur due to the proximity from the coast and moderate surrounding hills (Chen et al., 2008<sup>2</sup>). However, during stable weather conditions, prevailing wind directions and air mass origin at the sampling sites were similar for several days, hence the main source regions can be deduced for the sampling periods as presented in Table 2. The accuracy of this source region analysis suffers somewhat from the uncertainties of the back trajectories (Stohl and Seibert, 1998). Furthermore, unstable weather conditions resulted in a mixture of air masses. Such episodes, which were more frequent during the summer campaign, were identified by divergence of trajectories arriving at different heights. Due to these limitations, the main source regions summarized in Table 2 have to be regarded with some cautiousness. Typical 10-days back trajectories are shown in Figs. 1 and 2.

For the winter campaign, the air masses frequently arrived from western, northern, and eastern directions, transporting particulate matter from the Atlantic Ocean, Scandinavia north of Göteborg, and Central/Eastern Europe, respectively. Air masses from the Atlantic Ocean are likely to consist of rather clean marine air, while air masses from the eastern sector are likely to contribute significantly more to the aerosol loading at the two sites. As listed in Table 2, the samples collected during the periods 11–14 February and 14–18 February were influenced by Central/Eastern European air masses, whereas the sample collected from the 25 February–4 March was affected by air masses originating from Scandinavia. The sample collected in the intermediate period, 18–25 February, was influenced by both source regions. The summer campaign was characterized by westerly/south-westerly winds and changing cyclonic/anticyclonic conditions. Whereas air from the UK was transported over the North Sea or the coastal regions of Continental Europe to Denmark and Sweden during the first part of the period, the latter was dominated by local Scandinavian air masses. The periods 20–27 June and 27 June–4 July are characterized by westerly winds and by Scandinavian air, respectively. During the latter episode, the back trajectory calcula-

<sup>2</sup>Chen, D., Tang, L., and Achberger, C.: Synoptic and local meteorological conditions during Göte-2005 field campaign, in preparation, 2008.

tions showed that the air masses traveled close to ground shortly before the sampling site, suggesting a possible contribution from local emission sources.

### 3.2 Concentrations of carbonaceous particles

For both winter and summer campaigns, Fig. 3 shows TC, OC (separated for WINSOC and WSOC), and EC concentrations, while Table 3 presents corresponding TC concentrations and EC/TC as well as WSOC/OC ratios. During the winter campaign, the TC concentration ( $2.1\text{--}3.6\ \mu\text{g m}^{-3}$ ) at the urban site was approximately a factor of two higher compared to the rural site. The mean EC/TC ratio was only slightly higher at the urban site ( $0.30\pm 0.04$ ) compared to the rural site ( $0.24\pm 0.01$ ), whereas the WSOC/OC ratio was substantially higher at the rural site ( $0.61\pm 0.05$ ) compared to the urban site ( $0.48\pm 0.06$ ). For summer, the TC concentration at Femman was  $2.2\text{--}3.0\ \mu\text{g m}^{-3}$  and, therefore, comparable to the winter measurements. However, the EC/TC ratios were substantially lower during summer ( $0.17\pm 0.04$ ), whereas WSOC/OC ratios were similar for both seasons.

### 3.3 Results of $^{14}\text{C}$ analysis

Table 3 shows the modern carbon fraction ( $f_M$ , see Sect. 2.3) of EC, OC, WSOC and WINSOC. In winter,  $f_M(\text{EC})$  was substantially lower for the urban site ( $0.12\pm 0.03$ ) compared to the rural site ( $0.39\pm 0.05$ ), showing a higher relative contribution of fossil fuel sources to EC the urban site, which most likely is attributed to the influence of vehicular traffic as discussed in detail below. For the winter,  $^{14}\text{C}$  isotope signals of OC and its sub-fractions were identical within uncertainties between the both sites and average values for  $f_M(\text{OC})$ ,  $f_M(\text{WINSOC})$ , and  $f_M(\text{WSOC})$  were 0.70, 0.54, and 0.83, respectively. The average  $f_M(\text{EC})$  was similar during summer and winter for the urban site, however,  $f_M(\text{EC})$  showed higher variability during summer compared to winter. Concentrations of total EC and  $\text{EC}_{\text{fossil}}$  were higher by a factor of 2.4 and 3.1, respectively, at the urban site compared to the rural site, whereas  $\text{EC}_{\text{wood}}$  was about the same level

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at the two sites or even slightly increased at the rural station. This implies that the spatial variability of wood-burning emissions between the two sites (and, probably, in the whole region) during winter are moderate, whereas the contribution of fossil sources to EC strongly depend on the urban vs. rural site characteristics. During summer, EC concentrations were about half of that observed in winter level with small wood-burning contributions.

Figure 3 shows OC concentrations divided into water-soluble and water-insoluble organic carbon, which have been separated with respect to fossil and non-fossil sources. The WSOC fraction was dominated by non-fossil sources for all samples with an average non-fossil contribution of 70%, which did not differ significantly between the sites and seasons. WINSOC comprised a similar fossil and non-fossil content with average non-fossil contributions of 47%, 51%, and 56% ( $f_M$ : 0.52, 0.56, and 0.61) for urban/winter, rural/winter, and urban/summer conditions, respectively. Figure 3 indicates that the urban vs. rural difference during winter were more pronounced for WINSOC than for WSOC: whereas the urban/rural ratio of fossil WINSOC concentrations amounted 2.1, these ratios were 1.6, 1.4 and 1.3 for non-fossil WINSOC, fossil and non-fossil WSOC, respectively. Consequently, fossil WINSOC is the sub-fraction of OC that is influenced the most by the urban environment. For summer, the composition of OC was comparable to winter at the urban site with the exception that the water-soluble vs. water-insoluble distribution for fossil OC was shifted to WSOC for summer.

### 3.4 Levoglucosan measurements

The monosaccharide anhydrides levoglucosan, mannosan and galactosan were analysed, whilst only the mean concentration of levoglucosan is reported here (Table 3, Fig. 4). Levoglucosan was the most abundant isomer regardless of site and season, accounting for  $69\pm 4\%$  of the sum of the three isomers, followed by mannosan ( $23\pm 3\%$ ) and galactosan ( $9\pm 2\%$ ). The relative contribution of levoglucosan is in the lower range of what has been reported previously for European sites (e.g. Zdráhal et al., 2001;

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Pashynska et al., 2002; Yttri et al., 2005, 2008<sup>3</sup>; Szidat et al., 2006).

A mean concentration of  $62 \pm 30 \text{ ng m}^{-3}$  levoglucosan was observed at the urban site Femman in Göteborg during wintertime sampling (Table 3). Concurrent measurements of levoglucosan at the rural site Råö showed a somewhat lower mean concentration ( $35 \pm 20 \text{ ng m}^{-3}$ ). The mean concentration observed at the urban site in summer ( $17 \pm 11 \text{ ng m}^{-3}$ ) was more than three times lower than the concentrations observed in winter. Increased concentrations of levoglucosan in winter compared to summer has previously been reported by Zdrahal et al. (2001) for Ghent (Belgium), Szidat et al. (2006) for Zurich (Switzerland), and Yttri et al. (2008<sup>3</sup>) for Elverum (Norway), although more pronounced (up to factor of 20) than for Göteborg. The less evident seasonal variation observed in Göteborg is attributed to the low levels of levoglucosan in winter, while summertime levels are equal to those reported by Zdrahal et al. (2001) and Yttri et al. (2008<sup>3</sup>).

The mean concentration of levoglucosan observed at the urban site in Göteborg is in the lower range of what has been reported for other major (>500 000 inhabitants) cities in Scandinavia during wintertime sampling, i.e. Copenhagen (Oliveira et al., 2007), Helsinki (Saarnio et al., 2006; Sillanpää et al., 2005) and Oslo (Yttri et al., 2005, 2008<sup>3</sup>) (see Table 4). The difference is even larger when compared to small Scandinavian towns (<20 000 inhabitants), for which the mean concentrations of levoglucosan range between  $0.2\text{--}0.9 \mu\text{g m}^{-3}$  (Yttri et al., 2005; Hedberg et al., 2006; Glasius et al., 2008). Considerably higher concentrations of levoglucosan are reported even for cities in Continental and Southern Europe such as Amsterdam, Duisburg, Oporto and Prague (Sillanpää et al., 2005; Oliveira et al., 2007) (Table 4). It should be noted though that the levels presented by Sillanpää et al. (2005) were obtained during episodes with severe air pollution. Possible explanations to the lower levels of levoglucosan observed in Göteborg might be the state of combustion technology, the nature of the appliances used, and the extended use of district heating.

<sup>3</sup>Yttri, K. E., Dye, C., Braathen, O.-A., Simpson, D., and Steinnes, E.: Carbonaceous aerosols at urban influenced sites in Norway, Atmos. Chem. Phys. Discuss., submitted, 2008.

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A wide range of levoglucosan-to-OC emission ratios has been reported for various types of softwood and hardwood (Fine et al., 2002a; Simoneit et al., 1999). Further, levoglucosan-to-OC emission ratios have typically been established for US tree types (Fine et al., 2001, 2002a, b, 2004) using US appliances, which may not be applicable to Europe. Fine et al. (2002a) found that the levoglucosan-to-OC emission ratio ranged from 113–191 mg g<sup>-1</sup> for ten regions covering the entire US, when normalizing with respect to local tree types and appliances used. Thus, levoglucosan accounted for 14±2% of the fine OC emission. Emission ratios for European tree types are scarce. Puxbaum et al. (2007) reported a levoglucosan-to-OC-ratio (PM<sub>10</sub>) of 14% based on test burns of beech (7.9%) and spruce (20.6%). Considering these European studies (and accounting to some extent for the high values found in Hedberg et al., 2006), Simpson et al. (2007) suggested a central-estimate of 13%, ranging between 6.5 and 26%.

For Scandinavia, Yttri et al. (2008)<sup>3</sup> found levoglucosan/OC emission ratios of 8.9% (likely range 7.1–12%) for PM<sub>2.5</sub> and 6.7% (range 5.6–9.1%) for PM<sub>10</sub>, in an area almost exclusively influenced by residential wood burning. That study accounted for the positive sampling artifacts of OC and so likely produced higher levoglucosan/OC ratios than are applicable for our work. By allowing for this difference between both studies and considering other studies, we chose a levoglucosan/OC emission ratio of 10% (range 7.2–18%). This is equivalent to (OC/lev)<sub>wood</sub> of 10 (range 5.5–14), which is used in this work.

### 3.5 Source apportionment results

Table 5 gives concentrations of EC and OC distinguished into different sources. The determination of EC<sub>fossil</sub> and EC<sub>wood</sub> as well as OC<sub>fossil</sub> and OC<sub>nonfossil</sub> is based on <sup>14</sup>C analysis (see Sect. 2.5). For the separation of OC<sub>nonfossil</sub> into OC<sub>wood</sub> and OC<sub>bio</sub>, <sup>14</sup>C and levoglucosan measurements were used in combination. The uncertainties of these sources were calculated with LHS (Gelencsér et al., 2007) allowing for complex parameter combinations and asymmetric confidential bands. These ranges represent the

probability distribution (10th and 90th percentile) of that component's contribution, indicating the most-likely value for each component and its precision. LHS was especially helpful for the determination of  $OC_{wood}$ .  $^{14}C$  and levoglucosan enable independent estimations of this source. However, both approaches require OC-to-tracer emission ratios from laboratory studies, which often show high variabilities. In order to improve the reliability of  $OC_{wood}$ , results of both approaches were combined using LHS. The broad distributions of  $OC_{wood}$  (and  $OC_{bio}$  accordingly) originate mainly from uncertainties of the input parameters. Furthermore, considerable differences between the  $^{14}C$  and the levoglucosan approach occurred for a few samples: for 11–14 February at Femman and 14–18 February at Råö, 90% confidential bands of both approaches did not overlap. The reasons for this large difference remain unclear. Major uncertainties may derive from variable tracer-to-OC emission ratios for both, levoglucosan and  $^{14}C$ , due to spatially and temporarily changing burning and emission conditions. Furthermore, method-dependent differences between sites or seasons cannot be excluded as well, especially for the separation of OC and EC (Penner and Novakov, 1996; Szidat et al., 2004c), which is necessary for the estimation of  $OC_{wood}$  with  $^{14}C$ . Typical uncertainties (90% confidence) amount 20% for the separation of EC into  $EC_{fossil}$  and  $EC_{wood}$  as well as for OC into  $OC_{fossil}$  and  $OC_{nonfossil}$ , whereas they are higher for the determination of  $OC_{bio}$  and  $OC_{wood}$ : for  $OC_{wood}$ , uncertainties reach 30–50%, which result in uncertainties for  $OC_{bio}$  of 20% for summer and >30% for winter. For Råö, the uncertainty ranges are much larger than for Femman. Thus, the source allocation for Femman seems much more robust than that for Råö.

## 4 Discussion

### 4.1 Local and seasonal intercomparison

Source apportionment of the carbonaceous aerosol from Table 5 is summarized in Fig. 5 and compared to previous results from Zurich, Switzerland in Fig. 6. During the

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winter campaign, the urban impact is more pronounced for the urban site compared to the rural site, as seen from the higher carbonaceous aerosol concentration and a higher EC/TC ratio. Furthermore, the fossil impact is relatively larger at the urban site as shown by the EC fraction, which originates nearly exclusively from fossil sources.

5 The rural characteristics of the Råö site gives rise to a substantially higher relative importance of wood burning, which is indicated by the elevated contributions of  $OC_{wood}$  and  $EC_{wood}$  (Fig. 6). However, the absolute concentrations of wood burning aerosols seem to be elevated only slightly at the rural compared to the urban site, as shown by the nearly similar levels of  $EC_{wood}$  at both locations. The presence of  $OC_{bio}$  is  
10 consistent with model calculations suggesting that biogenic SOA can be formed in Scandinavia during winter (Simpson et al., 2007), even if both approaches, the  $^{14}C$  method and current modeling calculations, result in highly uncertain estimations of biogenic SOA concentrations for winter.

Comparing the results from the wintertime sampling in greater Göteborg with that reported for Zurich (Figs. 5 and 6), we found that the similarities was most pronounced  
15 for the rural site outside Göteborg rather than for the urban site. These similarities were related to the EC/TC ratios, fossil contributions, and  $EC_{wood}$ . We assume that the different histories of the air masses might cause this effect: whereas Göteborg is influenced by relatively clean air from marine or low-populated land areas, the air masses entering Zurich have traveled over moderately-populated regions with a rural  
20 character carrying aerosols which are accumulated over several days. Consequently, the local urban impact is better detectable at Göteborg.

$OC_{bio}$  is the dominating source both in greater Göteborg and in Zurich during summer. In contrast to winter, the significant existence of this fraction is unquestionable for  
25 summer. For Zurich, OC was shown to originate mainly from biogenic SOA with minor contributions from primary biogenic OC (Szidat et al., 2004a, b), and the model calculations for Nordic sites by Simpson et al. (2007) would suggest a similar preponderance of SOA also for the Göteborg region. As a consequence of this additional summertime OC source, EC/TC ratios are reduced compared to wintertime.



## 4.2 Influence of air mass origin

During the winter campaign, the first two samples (i.e., 11–14 and 14–18 February) experienced air from Central and Eastern Europe. These are characterized by higher values for  $f_M(\text{EC})$  and  $f_M(\text{WINSOC})$ , indicating a larger impact of EC and probably also of primary OC from wood burning. Aerosols in the last sample (25 February–4 March) were associated with air masses from Northern Scandinavia. This sample showed lower values for  $f_M(\text{EC})$  and  $f_M(\text{WINSOC})$ , reflecting the increased urban influence of local fossil emissions for EC and primary OC. On the other hand,  $f_M(\text{WSOC})$  is nearly on the contemporary level, which means that SOA is marginally formed from fossil VOCs. This observation indicates presence of biogenic SOA, which has already been suggested for Pittsburgh during winter by Subramanian et al. (2007). Other possible contemporary sources are quite unlikely for the last sample (25 February–4 March):

- a) the effect from wood burning is small as revealed by the low  $f_M(\text{EC})$ .
- b) in contrast to our observations, cooking emissions should be detectable by high values for  $f_M(\text{WINSOC})$  rather than for  $f_M(\text{WSOC})$ , as typical cooking tracers like fatty acids and cholesterol (Subramanian et al., 2007) are water insoluble; due to the vicinity of many restaurants at the Femman site, however, contributions from cooking cannot be excluded totally.

For the summer campaign, the second sample (20–27 June) was dominated by westerly/south-westerly winds carrying air masses from the UK and the North Sea over Denmark to Göteborg. For this sample, unusual high  $f_M(\text{EC})$  values were determined,  $\approx 2.5$  times higher than for average summer conditions in Zurich (Szidat et al., 2004a). According to MODIS fire maps (<http://rapidfire.sci.gsfc.nasa.gov/firemaps>), hardly any wild fire activity occurred in the source region in the end of June 2006, thus the reason for the high  $f_M(\text{EC})$  values remain unexplained. The last summer sample (27 June–4 July) experienced air masses from Central Scandinavia and the Baltic Sea region. Here, EC was found to be nearly exclusively from fossil sources, which was observed

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for Zurich as well (Szidat et al., 2004a). Moreover,  $^{14}\text{C}$  levels of OC, WINSOC, and WSOC are lower as well compared to the other two summer samples, reflecting the importance of the local urban emissions from fossil sources for this sample. In particular, the  $\sim 40\%$  fossil fraction of WSOC – a proxy for fossil SOA – is remarkable, as it is twice as high as for Zurich (Szidat et al., 2004a, 2006).

## 5 Conclusions

Particulate matter was collected at the urban site Femman in Göteborg, Sweden, for a summer and a winter period. Concurrent measurements were performed at the rural site, Råö, 35 km south of Göteborg, during the winter period. The carbonaceous particle fractions EC, OC, WINSOC, and WSOC were sequentially separated from the filters by water extraction and oxidation.  $^{14}\text{C}/^{12}\text{C}$  isotopic ratios were determined as fractions of modern carbon ( $f_M$ ) and fossil vs. non-fossil sources were apportioned for every fraction. Separate determination of the wood burning aerosol tracer compound levoglucosan made it possible to refine the contribution of this source using LHS calculations. The main results are:

- The urban site was considerably influenced by local urban sources in winter, as seen from the high relative contribution of  $\text{EC}_{\text{fossil}}$  to EC ( $f_M(\text{EC})=0.12$ ). The corresponding value for Råö (0.39) was more than three times less than that observed at the urban site, and comparable to that reported for an urban site in Zurich (0.31), using the same methodology. A low impact of wood burning was evident for the urban site.
- In winter, the mean concentration of  $\text{EC}_{\text{wood}}$  at the rural site ( $0.15 \mu\text{g m}^{-3}$ ) was slightly higher than that of the urban site ( $0.09 \mu\text{g m}^{-3}$ ), whereas the concentrations of  $\text{EC}_{\text{fossil}}$  was considerably lower ( $0.30 \mu\text{g m}^{-3}$  at the rural site compared to  $0.81 \mu\text{g m}^{-3}$  at the urban site). This implies that the spatial variability of wood burning aerosols is moderate.

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- For winter, air masses originating from Scandinavia ( $f_M(\text{EC})=0.11$ ) were characterized by high  $f_M$  values for WSOC (0.98) and low  $f_M$  values for EC (0.11), which suggests the abundance of biogenic SOA even in winter.
- For summer,  $f_M(\text{WSOC})$  indicated a relatively high fossil contribution to SOA (~40%) for aerosols at the urban site, which was of regional origin. This is approximately twice as high as for Zurich. The impact of wood burning was minor during the summer.

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**Table 1.** Conditions of aerosol sampling.

Campaign	Winter, urban	Winter, rural	Summer, urban
Sampling period	11 February–4 March 2005	14 February–25 February 2005	13 June–4 July 2006
Site	Femman	Råö	Femman
Position	57°42'N, 11°57'E, 20 m a.s.l.	57°24'N, 11°55'E, 10 m a.s.l.	See winter, urban
Location	Downtown Göteborg (600 000 inhabitants), rooftop position	2 km SW of Råö, ground position	See winter, urban
Vicinity	Shopping, residential, and industrial areas	Forest, heath, sea shore	See winter, urban
Sampling system ( <sup>14</sup> C)	Sierra-Andersen GMW HiVol Air Sampler Flow: 1130 L min <sup>-1</sup> Inlet: PM <sub>10</sub>	Anderson 65–800 Two-Stage HiVol Cascade Impactor Flow: 480 L min <sup>-1</sup> Inlet: PM <sub>2.5</sub>	Anderson 65–800 Two-Stage HiVol Cascade Impactor Flow: 480 L min <sup>-1</sup> Inlet: PM <sub>2.5</sub>
Sampling system (levoglucosan)	Derenda LVS3 Flow: 38 L min <sup>-1</sup> Inlet: PM <sub>2.5</sub>	Identical to <sup>14</sup> C	Identical to <sup>14</sup> C

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**Table 2.** Source regions of air masses reaching Råö according to FLEXTRA ten-days back trajectories (Stohl and Seibert, 1998). Typical trajectories are shown in Figs. 1 and 2.

Sample	Period	Wind direction	Source regions
11–14 Feb 2005	11–12 Feb	W/NW	Greenland, Atlantic Ocean
	13–14 Feb	E/NE	Central Europe, Baltic states
14–18 Feb 2005	14–17 Feb	NE	Ukraine, Russia, Finland
	18 Feb	W	Atlantic Ocean
18–25 Feb 2005	18–19 Feb	W/NW	Atlantic Ocean
	20–21 Feb	N/NE	Arctic Ocean, Scandinavia
	21–25 Feb	E/NE	Ukraine, Russia, Finland
25 Feb–4 Mar 2005	25 Feb–4 Mar	N/NE	Arctic Ocean, Scandinavia
13–20 Jun 2006	13–14 Jun	W/SW	UK, North Sea, Denmark
	15–17 Jun	variable	Scandinavia
	18–20 Jun	W/SW	UK, North Sea, Denmark
20–27 Jun 2006	20–27 Jun	W/SW	UK, North Sea, Denmark
27 Jun–4 Jul 2006	27 Jun–2 Jul	variable	Scandinavia
	2–4 Jul	S/SE	Baltic Sea (coastal and open sea)

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**Table 3.** TC concentrations, EC/TC as well as WSOC/OC ratios,  $f_M$  determinations and levoglucosan (lev) concentrations at the urban and the rural site during winter 2005 and summer 2006. All values are presented with combined measurement uncertainties ( $1\sigma$ ).

Sample	TC [ $\mu\text{g m}^{-3}$ ]	EC/TC	WSOC/OC	$f_M(\text{EC})$	$f_M(\text{OC})$	$f_M(\text{WINSOC})$	$f_M(\text{WSOC})$	Lev [ $\mu\text{g m}^{-3}$ ]
Winter, urban								
11–14 Feb 2005	2.1±0.1	0.28±0.04	0.44±0.06	0.14±0.03	0.74±0.02	0.67±0.02	0.82±0.05	0.06 <sup>a</sup>
14–18 Feb 2005 <sup>b</sup>	3.1±0.2	0.31±0.04	0.55±0.06	0.15±0.02	0.67±0.02	0.52±0.02	0.79±0.05	0.05 <sup>a</sup>
18–25 Feb 2005 <sup>b</sup>	3.2±0.2	0.35±0.05	0.51±0.06	0.08±0.01	0.63±0.02	0.46±0.01	0.80±0.05	0.06 <sup>a</sup>
25 Feb–4 Mar 2005	3.6±0.2	0.27±0.03	0.42±0.06	0.11±0.01	0.67±0.02	0.45±0.01	0.98±0.08	0.07 <sup>a</sup>
Average	3.0±0.7	0.30±0.04	0.48±0.06	0.12±0.03	0.68±0.04	0.52±0.10	0.85±0.09	0.06±0.01
Winter, rural								
14–18 Feb 2005 <sup>b</sup>	1.8±0.1	0.24±0.03	0.65±0.06	0.42±0.06	0.68±0.02	0.55±0.04	0.75±0.05	0.02
18–25 Feb 2005 <sup>b</sup>	1.9±0.1	0.25±0.03	0.57±0.06	0.35±0.03	0.74±0.02	0.57±0.02	0.87±0.04	0.05
Average	1.8±0.1	0.24±0.01	0.61±0.05	0.39±0.05	0.71±0.04	0.56±0.02	0.81±0.08	0.04±0.02
Summer, urban								
13–20 Jun 2006	2.9±0.2	0.13±0.02	0.43±0.06	0.13±0.02	0.73±0.01	0.68±0.01	0.80±0.03	0.01
20–27 Jun 2006	2.2±0.1	0.17±0.02	0.62±0.06	0.17±0.03	0.78±0.01	0.64±0.01	0.87±0.03	0.03
27 Jun–4 Jul 2006	3.0±0.2	0.20±0.02	0.54±0.06	0.05±0.02	0.59±0.01	0.52±0.01	0.65±0.02	0.01
Average	2.7±0.4	0.17±0.04	0.53±0.10	0.12±0.06	0.70±0.10	0.61±0.08	0.78±0.11	0.02±0.01

<sup>a</sup> Averaged from daily measurements, which ended on 2 March (see Fig. 4).

<sup>b</sup> Note that sampling was performed simultaneously at both sites.

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**Table 4.** Concentrations ( $\mu\text{g m}^{-3}$ ) of levoglucosan (lev) and monosaccharide anhydrides ( $\Sigma\text{MA}$ , i.e. sum of levoglucosan, mannosan and galactosan) reported for European urban areas during wintertime.

Study	Site	Category	<i>n</i>	Date	Size	Lev	$\Sigma\text{MA}$
Zdrahal et al. (2002)	Ghent (Belgium)	Urban background	8	1998 (Jan–Mar)	PM <sub>10</sub>	0.48	0.56
Paschynska et al. (2002)	Ghent (Belgium)	Urban background	9	2000/01 (Nov–Mar)	PM <sub>10</sub>	0.42	0.51
Yttri et al. (2005)	Oslo (Norway)	Urban background	24	2001 (Nov–Dec)	PM <sub>10</sub>	0.17±0.15	0.21±0.19
	Elverum (Norway)	Urban background	25	2002 (Jan–Mar)	PM <sub>10</sub>	0.41±0.22	0.53±0.28
Sillanpää et al. (2005); Saarikoski et al. (2008)	Amsterdam (NL)	Urban background	14	2003 (Jan–Mar)	PM <sub>2.5</sub>	–	0.36±0.25
	Duisburg (Germany)	Urban background	14	2002 (Oct–Nov)	PM <sub>2.5</sub>	–	0.23±0.13
	Helsinki (Finland)	Urban background	15	2002 (Mar–May)	PM <sub>2.5</sub>	–	0.88±0.07
	Prague (CZ)	Urban background	13	2002/03 (Nov–Jan)	PM <sub>2.5</sub>	–	1.32±0.50
Hedberg et al. (2006)	Lycksåle (Sweden)	Urban background	15	2002 (Jan–Mar)	PM <sub>10</sub>	0.90	–
Saarnio et al. (2006)	Helsinki (Finland)	Urban background	9	2004 (Jan–Feb)	PM <sub>2.5</sub>	–	0.25±0.14
	Kotka (Finland)	Urban background	20	2005/06 (Nov–May)	PM <sub>2.5</sub>	–	0.07±0.04
Szidat et al. (2006)	Zurich (Switzerland)	Urban background	2	2003 (Feb)	PM <sub>10</sub>	0.62±0.16	–
Sandradewi et al. (2008b)	Zurich (Switzerland)	Urban background	7	2006 (Jan)	PM <sub>1</sub>	0.31±0.16	0.41±0.21
Oliveira et al. (2007)	Oporto (Portugal)	Urban background	28	2003 (Nov–Dec)	PM <sub>10</sub>	0.12±0.08	–
	Roadside		28	2003 (Nov–Dec)	PM <sub>10</sub>	0.17±0.10	–
	Copenhagen (DK)	Urban background	27	2004 (Feb–Mar)	PM <sub>10</sub>	0.06±0.04	–
	Roadside		27	2004 (Feb–Mar)	PM <sub>10</sub>	0.11±0.06	–
Glasius et al. (2008)	Vindinge (Denmark)	Urban background	10	2005 (Feb–Apr)	PM <sub>2.5</sub>	0.17±0.09	–
	Residential		10	2005 (Feb–Apr)	PM <sub>2.5</sub>	0.31±0.24	–
This study	Göteborg	Urban background	22	2005 (Feb–Mar)	PM <sub>2.5</sub>	0.06±0.03	0.09±0.05
	Råö	Rural	2	2005 (Feb–Mar)	PM <sub>10</sub>	0.04±0.02	–

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**Table 5.** Concentrations ( $\mu\text{g m}^{-3}$ ) of different carbonaceous particle fractions. Distributions from Latin-hypercube sampling (LHS) calculations (Gelencsér et al., 2007) are given as medians with 10th and 90th percentiles.

Sample	EC <sub>fossil</sub>	EC <sub>wood</sub>	OC <sub>fossil</sub>	OC <sub>wood</sub>	OC <sub>bio</sub>
Winter, urban					
11–14 Feb 2005	0.51 (0.40, 0.61)	0.07 (0.04, 0.08)	0.53 (0.40, 0.65)	0.53 (0.38, 0.67)	0.46 (0.23, 0.67)
14–18 Feb 2005*	0.83 (0.65, 0.99)	0.12 (0.09, 0.16)	0.88 (0.68, 1.05)	0.65 (0.43, 0.84)	0.61 (0.28, 0.90)
18–25 Feb 2005*	1.02 (0.80, 1.22)	0.08 (0.03, 0.10)	0.94 (0.77, 1.09)	0.55 (0.38, 0.67)	0.61 (0.32, 0.86)
25 Feb–4 Mar 2005	0.88 (0.68, 1.04)	0.09 (0.07, 0.14)	1.07 (0.83, 1.26)	0.65 (0.47, 0.83)	0.91 (0.61, 1.19)
Winter, rural					
14–18 Feb 2005*	0.27 (0.20, 0.32)	0.15 (0.11, 0.18)	0.55 (0.43, 0.65)	0.49 (0.23, 0.72)	0.34 (0.07, 0.59)
18–25 Feb 2005*	0.33 (0.25, 0.38)	0.14 (0.10, 0.17)	0.50 (0.38, 0.61)	0.65 (0.44, 0.82)	0.29 (0.06, 0.51)
Summer, urban					
13–20 Jun 2006	0.34 (0.26, 0.38)	0.04 (0.03, 0.09)	0.87 (0.64, 1.07)	0.19 (0.09, 0.26)	1.47 (1.25, 1.68)
20–27 Jun 2006	0.32 (0.24, 0.37)	0.06 (0.02, 0.07)	0.56 (0.37, 0.70)	0.33 (0.22, 0.40)	0.94 (0.75, 1.12)
27 Jun–4 Jul 2006	0.57 (0.45, 0.66)	0.02 (0.00, 0.06)	1.13 (0.93, 1.29)	0.14 (0.09, 0.18)	1.14 (0.96, 1.29)

\* Note that sampling was performed simultaneously at both sites.

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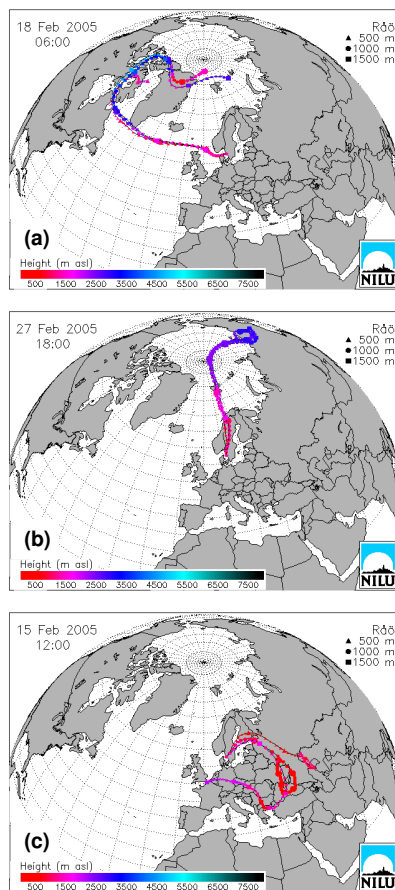
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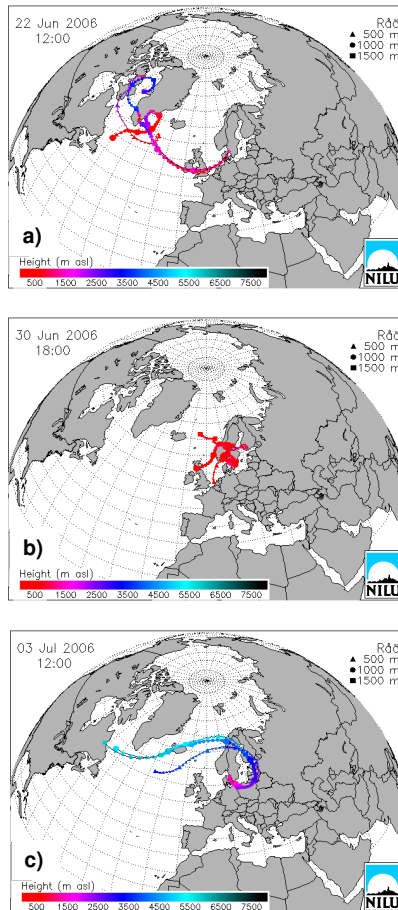


**Fig. 1.** Examples of FLEXTRA ten-days back trajectories (Stohl and Seibert, 1998) for air masses reaching Rådö, which originate from the North Atlantic Ocean (a), North Scandinavia (b), and Eastern Europe (c) during winter 2005.

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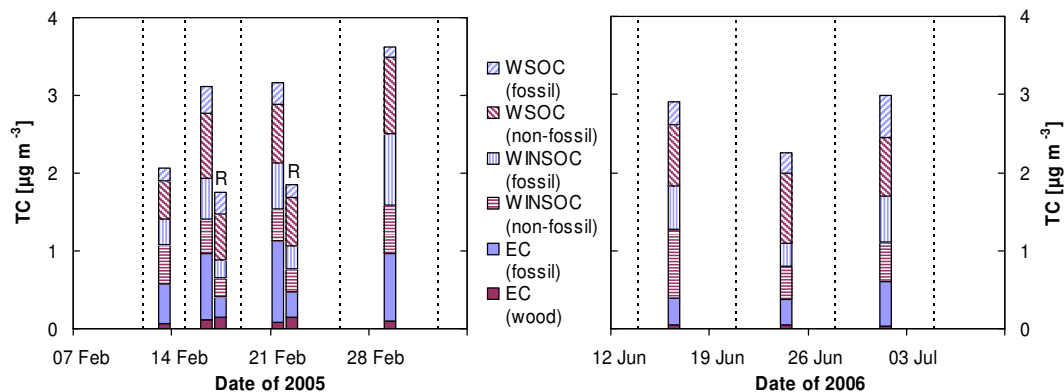


**Fig. 2.** Examples of FLEXTRA ten-days back trajectories (Stohl and Seibert, 1998) for air masses reaching Råö, which originate from UK/North Sea/Denmark **(a)**, Scandinavia **(b)**, and the Baltic Sea **(c)** during summer 2006.

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**Fig. 3.** Contributions of fossil and non-fossil sources to EC and OC during winter 2005 (left) and summer 2006 (right). Results from the rural site Råö are only available for winter 2005 and are indicated by “R”. Horizontal lines mark beginning and end of aerosol sampling.

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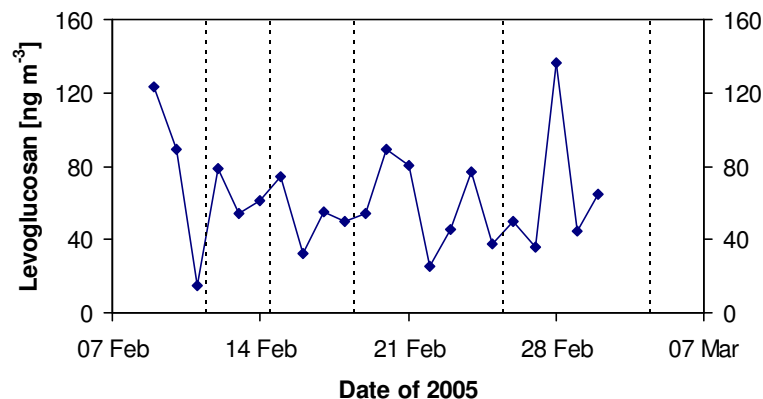
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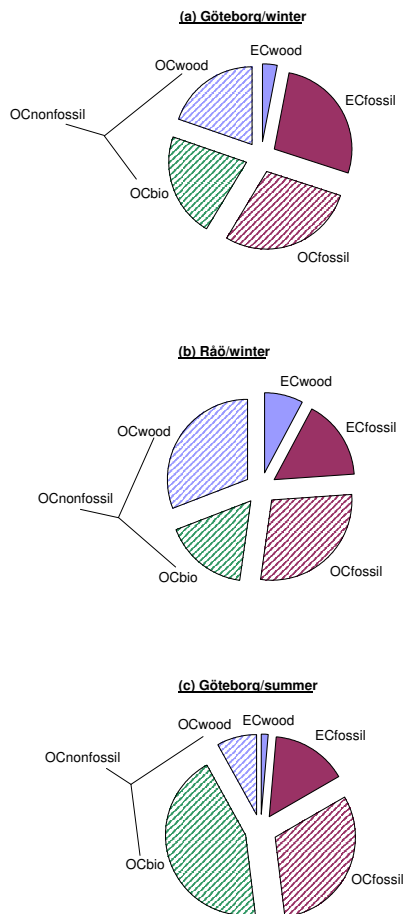


**Fig. 4.** 24-h mean concentrations of levoglucosan for the urban site during the winter campaign 2005. The horizontal lines mark the time periods during which aerosol filter samples were collected for subsequent <sup>14</sup>C analysis (see Fig. 3).

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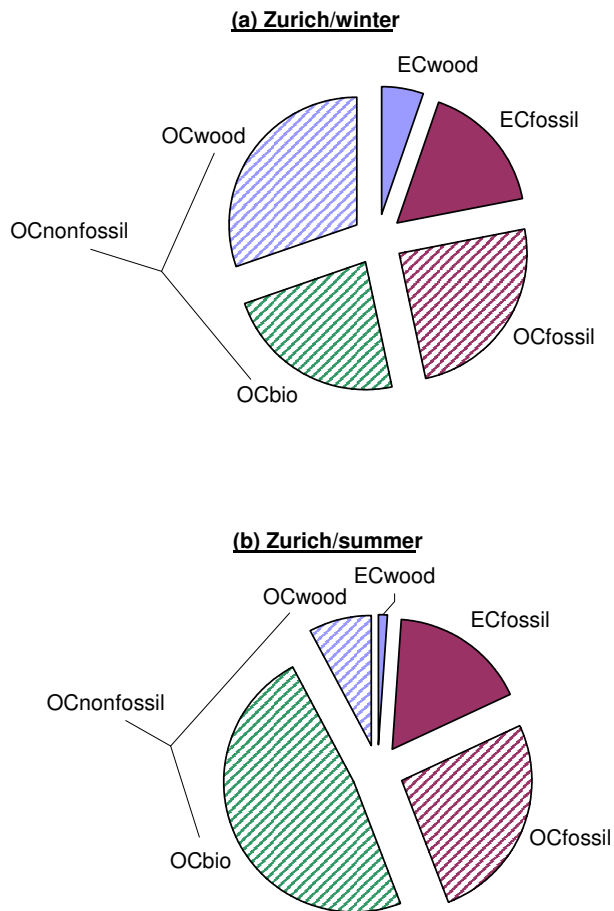


**Fig. 5.** Relative contributions of EC<sub>fossil</sub>, EC<sub>wood</sub>, OC<sub>fossil</sub>, and OC<sub>nonfossil</sub> (differentiated into OC<sub>wood</sub> and OC<sub>biogenic</sub>) to TC for **(a)** urban site Göteborg/winter, **(b)** rural site Råö/winter, and **(c)** urban site Göteborg/summer.

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**Fig. 6.** Relative contributions of  $EC_{\text{fossil}}$ ,  $EC_{\text{wood}}$ ,  $OC_{\text{fossil}}$ , and  $OC_{\text{nonfossil}}$  (differentiated into  $OC_{\text{wood}}$  and  $OC_{\text{biogenic}}$ ) to TC according to Szidat et al. (2006) for **(a)** Zurich/winter (February 2003), **(b)** Zurich/summer (August 2002).

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